

## Supporting Information

### A molecular crowding thermo-switchable chiral G-quartet hydrogel with circularly polarized luminescence property

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#### Experimental Section

##### Materials:

Guanosine 5'-monophosphate (GMP,  $\geq 99$ ), Guanosine (Gua,  $\geq 99$ ), thioflavin T (ThT, 65-75%) were purchased from Aladin and used without further purification. Polyethylene glycol 200 (PEG 200) was purchased from Sigma-Aldrich and used without further purification. ThT is quantified by UV spectrometer, the extinction coefficients of ThT is  $36000 \text{ M}^{-1} \text{ cm}^{-1}$  at 412 nm. All stock solutions are prepared with ultrapure water (18.2 M $\Omega$ , Milli-Q, Millipore).

##### Methods :

###### 1 · Preparation of GMP/Gua hydrogels:

GMP · Gua were dissolved in ultrapure water to prepare a higher concentration stock solution. GMP (50mM) and Gua (50mM) were dissolved in PEG 200 (50 Vol%). Due to the poor solubility of Gua in water, Gua stock solution should be pipetted several times before added to the mixture. Then the mixture was heated in water bath ( $\geq 70^\circ\text{C}$ ) until totally homogenised. After cooling to room temperature, white hydrogel formed. The chirality of the hydrogel depends on the speed of gelling process. When the hot homogeneous solution is cooled in an ice water bath, a right-handed hydrogel will be obtained, but cooling under annealing conditions will produce a left-handed hydrogel.

###### 2 · Preparation of GMP/Gua-ThT hydrogels:

GMP (50mM) and Gua (50mM) were dissolved in PEG 200 (50 Vol%). The mixture was heated in water bath ( $\geq 70^\circ\text{C}$ ) until totally homogenised. ThT (1.5mM) was added into the transparent mixed solution. The method of controlling chirality is the same as preparation of GMP/Gua hydrogels.

##### Measurements:

###### 1 · CD spectra measurement

CD spectra were measured at  $20^\circ\text{C}$  on a JASCO J-1500 spectrometer. Samples were sandwiched between two 1mm quartz plates. CPL spectra were carried out on a JASCO CPL-300 spectrometer at  $20^\circ\text{C}$  using a quartz cuvette with 0.1 mm path length. Each spectrum was an average of three measurements.

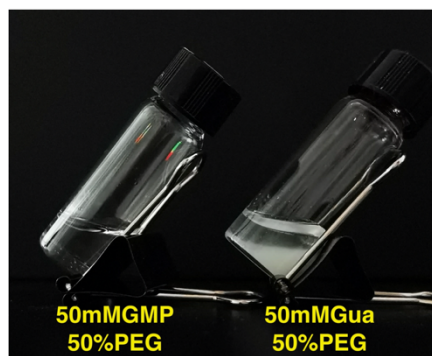
###### 2 · Thermal melting experiments

Thermal melting experiments were performed in a LAMBDA 750 spectrophotometer, the

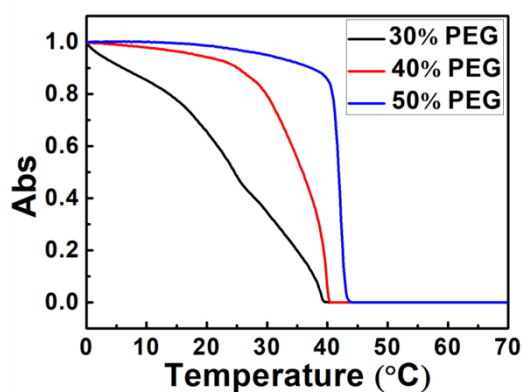
heating rate is 1°C/min at 420nm with a cuvette which optical path length is 1mm.

### 3 · Scanning electron microscopy and X-ray diffraction analyses

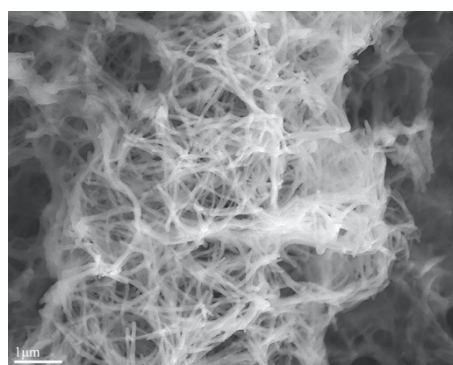
Scanning electron microscopy (SEM) was conducted on a GEMINI 300 field emission scanning microscope with SE2 detector. X-ray diffraction (XRD) analyses were carried out on a Bruker-D2 PHASE X-ray diffraction system and operating in a  $2\theta$  range from 10° to 80°. Samples were all freeze-dried before XRD analyses.



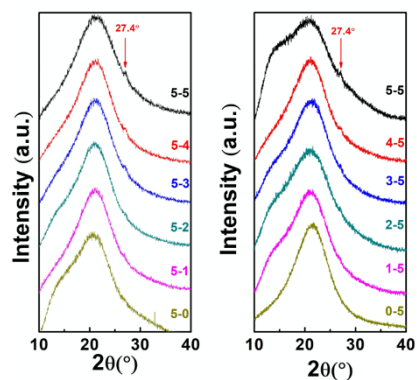
**Figure S1** 50mM GMP/Gua in 50%PEG.



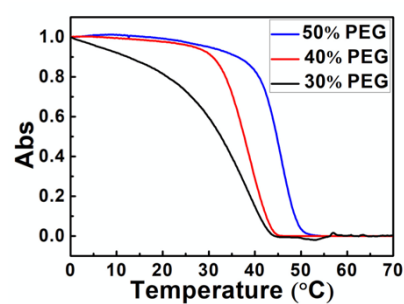
**Figure S2** UV melting spectra of GMP-Gua-PEG hydrogels of 30%, 40%, 50% PEG in 420nm.



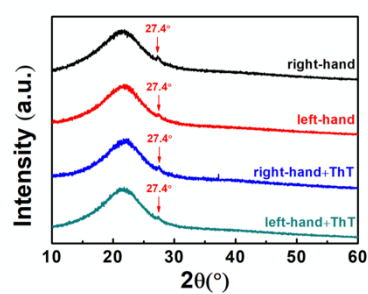
**Figure S3** SEM image of GMP-Gua-PEG hydrogel.



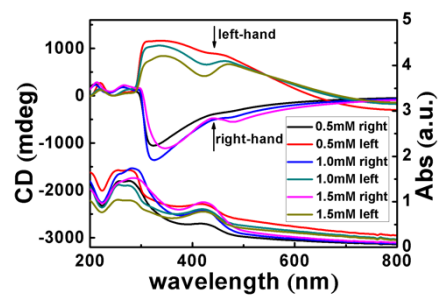
**Figure S4 C.** XRD spectra of GMP-Gua-PEG hydrogels in 50%PEG. The concentration of GMP is fixed at 50mM and the ratio of GMP to Gua is 5-5, 5-4, 5-3, 5-2,5-1. The concentration of Gua is fixed at 50mM and the ratio of GMP to Gua is 5-5, 5-4, 5-3, 5-2,5-1, 5-0. The concentration of Gua is fixed at 50mM and the ratio of GMP to Gua is 5-5, 5-4, 5-3, 5-2,5-1. The concentration of Gua is fixed at 50mM and the ratio of GMP to Gua is 5-5, 4-5, 3-5, 2-5, 1-5, 0-5.



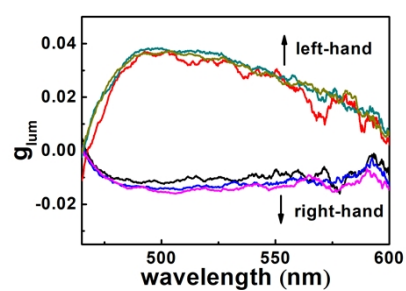
**Figure S5** UV melting spectra of GMP-Gua-PEG hydrogels of 30%, 40%, 50% PEG in 420nm.



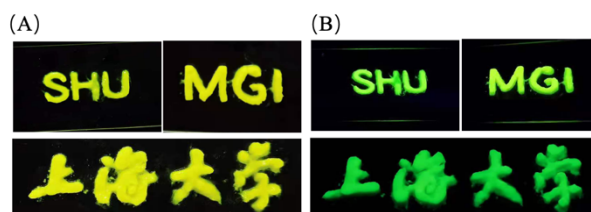
**Figure S6** XRD spectra of GMP-Gua-PEG hydrogels with and without ThT and formed in fast and slow speed.



**Figure S7** CD spectra of GMP-Gua-PEG hydrogels in opposite chirality with ThT from 0.5 to 1.5 mM. The signal intensity in 492nm increased with the concentration of ThT.



**Figure S8**  $g_{lum}$  of GMP-Gua-PEG hydrogels with ThT.



**Figure S9** GMP-Gua-PEG hydrogel with ThT under (A) daylight and (B) 365nm UV light.